

10/585,793

Remarks**A. Basis for claim amendments**

Basis for new dependent claims 12-14 includes page 8, lines 4-9 of the specification, reproduced below:

[0020] Incidentally, acrylic acid produced by the production process of the present invention can be industrially further subjected to publicly known purification methods (for example, processed by steps such as a step of collecting acrylic acid as a solution by using water or a solvent, a distillation step for removing low- and high-boiling-point materials from the resultant solution containing acrylic acid, or a crystallization step for purifying acrylic acid by crystallizing it) to thus provide acrylic acid as a product.

Basis for new independent claim 15 includes independent claim 1 and dependent claims 6, 7, and 8. Further basis for new independent claim 15 includes page 3, lines 1-13 of the specification, reproduced below:

[0009] The present invention uses glycerol as a raw material. The glycerol to be subjected to the dehydration reaction as a raw material may be 100% pure glycerol, or may be an aqueous glycerol solution which is a mixture of glycerol and water. Glycerol and the aqueous glycerol solution are recovered in hydrolysis of various oils-and-fats or from waste fluids in soap production and are therefore industrially easily available. In addition, glycerol is expected to be generated in abundance as a by-product in production of a bio-diesel fuel as a renewable fuel by hydrolysis of vegetable oils in the future, and effective use thereof is desired. The present invention provides a process for producing acrylic acid as a useful chemical from glycerol which is such an easily available and renewable raw material, and this process is also a process for providing acrylic acid of which the source of carbon is carbon dioxide fixed by plants and which substantially does not lead to the increase of carbon dioxide in the air even when incinerated.

(10450.DOC) (Amendment and Remarks--page 6 of 19)

10/585,793

Basis for new dependent claims 16-17 includes old dependent claim 10 (presently canceled). Further basis for new dependent claims 16-17 includes page 8, lines 4-12 of the specification, reproduced below:

[0020] Incidentally, acrylic acid produced by the production process of the present invention can be industrially further subjected to publicly known purification methods (for example, processed by steps such as a step of collecting acrylic acid as a solution by using water or a solvent, a distillation step for removing low- and high-boiling-point materials from the resultant solution containing acrylic acid, or a crystallization step for purifying acrylic acid by crystallizing it) to thus provide acrylic acid as a product. Then, this product can be used to produce, for example, polyacrylic acids (salts) as water-soluble polymers or water-absorbent resins, by publicly known polymerization methods such as thermal polymerization methods and photopolymerization methods.

B. The Office Action

There are three rejections pending:

1. Claims 1 and 3-6 are rejected under 35 U.S.C. 103(a) as being unpatentable over Unverricht et al. (US 6,525,217), in view of Hoyt (US 2,558,520);
2. Claims 1 and 3 are rejected under 35 U.S.C. 103(a) as being unpatentable over Unverricht et al. (US 6,525,217), in view of Hoyt (US 2,558,520), further in view of Unverricht et al. (US 6,403,829); and
3. Claims 1 and 5 are rejected under 35 U.S.C. 103(a) as being unpatentable over Unverricht et al. (US 6,525,217), in view of Hoyt (US 2,558,520), further in view of Uchida et al. (US 4,871,700).

10/585,793

C. Applicant's discussionC.1. Claimed difference is present

Claim 1 is as follows:

1. (previously presented) A process for producing acrylic acid, comprising the steps of:
 - a) vaporizing a raw material comprising an aqueous glycerol solution to generate a first gas, wherein said aqueous glycerol solution has a water content of not more than 20% by weight;
 - b) applying a dehydration reaction to glycerol in a gas phase that includes said first gas; and then
 - c) applying a gas phase oxidation reaction to a gaseous reaction product formed by the dehydration reaction to obtain said acrylic acid.

Claim 1 positively recites that the dehydration reaction is conducted in a gas phase. However, in Hoyt, the dehydration reaction is conducted in a liquid phase (in an inert hydrocarbon liquid). Please see, for instance, claim 1 of Hoyt.

It is therefore respectfully submitted that independent claim 1 is allowable over each of the above noted three rejections on the basis of this claimed difference.

C.2. There is no teaching, suggestion or motivation to combine Unverricht et al. with Hoyt

There are a number of reasons why there is no teaching, suggestion or motivation to combine Unverricht et al. with Hoyt. These reasons are set out below.

C.2.a. The simple liquid phase process of Hoyt results in the generation of tar

A simple liquid phase process results in the generation of a tar and stagnation and accumulation of high-boiling-point impurities in a system and is therefore not suitable for industrial continuous

10/585,793

production. The problem of the generation of tar is acknowledged in Hoyt, and Hoyt does not describe eliminating the generation of tar but simply reducing it in column 2, lines 45-50:

It is essential in practicing the present method that the acid or anhydride be supported upon a carrier, since if the acid or anhydride is not so supported the yield of acrolein is poor and excessive tar formation results.

The Hoyt process may be expected to decrease the tar that is generated, but cannot be expected to entirely exclude the generation of tar.

The Hoyt patent also discloses in column 3, lines 6-11 the following:

All of the aforementioned liquids have the necessary characteristics for use in the method of the present invention, viz., a high boiling point, stability at the reaction temperature, and non-reactivity with the glycerol or its decomposition products.

However, even if a solvent does not react with glycerol or a product therefrom, a by-product having a high boiling point which stagnates in a system may react with glycerol or a product therefrom such as acrolein, thus causing the formation of tar or the reduction of yield.

C.2.b. Primary reference does not teach starting material; secondary reference is cited to cure primary reference

Applicant's first step is:

- glycerol to a gaseous reaction product (acrolein).

Applicant's second step is:

- gaseous reaction product (acrolein) to acrylic acid.

The Patent Office employs Unverricht et al. '217 (and its teaching of the petroleum product of propene) as the primary reference against applicant's first and second

10/585,793

steps. The Patent Office then employs the secondary reference Hoyt (and its teaching of glycerol) to cure the first step.

"Starting from propene" is an indispensable part of the Unverricht et al. process. One skilled in the art would not destroy an indispensable part of the primary reference to employ the combination of Unverricht et al. '217 (starting material of propene) and Hoyt (starting material glycerol).

C.2.c. A substance diffuses at a greater speed in a gas phase than a liquid phase; therefore a liquid phase is unlike a gas phase

In the interview of May 20, 2008, the Examiners pointed out a portion of the top of column 2 of the Hoyt reference. This portion reads: "The temperature at the top of the fractionating column was maintained at a point such that vaporized oil was returned to the container."

As pointed out by the Examiners in the interview, a part of the solvent may be vaporized in Hoyt. However, the reaction itself takes place in a liquid phase.

It is widely known to a skilled artisan that there is a great difference between a liquid phase and a gas phase as to the speed at which a substance diffuses in a reactor, so that there is also a great difference as to necessary properties of such as catalysts. Therefore, except for only a few examples, a replacement of a liquid phase reaction by a gas phase reaction or otherwise a replacement of a gas phase reaction by a liquid phase reaction is usually not carried out.

10/585,793

C.2.d. Taking to the next step the argument of the Examiners that it is relevant that Hoyt discloses a gas

As indicated in the section above, the Examiners assert that Hoyt is applicable for its disclosure as to "The temperature at the top of the fractionating column was maintained at a point such that vaporized oil was returned to the container." This disclosure of Hoyt is taken a step further.

Again, Hoyt discloses that a gas of a hydrocarbon solvent is generated. However, if this hydrocarbon solvent gas came into a second reactor (where a gas phase oxidation reaction of acrolein to acrylic acid is carried out), then there would be problems as to:

1. the hydrocarbon solvent gas being adsorbed into or onto a catalyst, thereby deactivating the catalyst or clogging the reaction tube, or
2. the hydrocarbon solvent gas undergoing combustion to consume oxygen, thereby causing a shortage of oxygen necessary for the oxidation reaction of acrolein, or
3. the combustion of the hydrocarbon solvent gas causing abnormal heat generation, thereby damaging the catalyst or reactor, or
4. the hydrocarbon solvent gas mingling into a formed acrylic acid gas, thereby damaging the purification step of acrylic acid.

C.2.e. Hoyt discloses octadecane and Unverricht discloses oxygen; the product of such a combination would be ignition

The Hoyt reference discloses the employment of "octadecane" in column 3, line 4. Octadecane is known to have a flash point of about 165 °C.

10/585,793

Unverricht '217 discloses, as for example in the abstract, that when acrylic acid is produced from acrolein, the production is carried out by a gas phase oxidation reaction under the coexistence of oxygen.

If a hydrocarbon solvent such as octadecane is mixed with oxygen, ignition is a likely result. This teaches away from the combination of Hoyt and Unverricht '217.

Therefore, the combination of Hoyt with Unverricht '217 needs complicated arrangements such as: the dehydration reaction of glycerol being carried out in the absence of oxygen and, after the mingling hydrocarbon solvent gas has been completely removed, acrylic acid being produced from an acrolein gas having been mixed with an oxygen-containing gas. Thus, the simple combination of Hoyt with Unverricht '217 cannot industrially be carried out.

C.2.f. Summary

In light of the reasons discussed in the above sections, it is respectfully submitted that there is no teaching, suggestion or motivation for combining Hoyt with Unverricht '217.

C.3. Unexpected advantages

Generally, this case relates to the steps of (1) acrolein being obtained from glycerol and (2) acrylic acid being obtained from acrolein.

In the first step cited above, ally alcohol is formed in the dehydration reaction of glycerol in a gas phase. Then, in the second step cited above, the formed ally alcohol is converted into acrylic acid. This formation and conversion of ally alcohol is presently thought to be one of reasons for the high yield of acrylic acid in the present invention. This formation and conversion mechanism was

10/505,793

unknown by the inventors at the time of the convention priority date (January 30, 2004) of this case.

If acrolein is prepared from glycerol in the way of the combination of Hoyt and Unverricht and thereafter used to produce acrylic acid, then at least a part of the above ally alcohol is unfavorably removed, so that the total yield of acrylic acid from glycerol is unfavorably lessened. However, if the product of the dehydration reaction of glycerol is directly subjected to the oxidation reaction as in the present invention, then acrylic acid is efficiently obtained.

C.4. New dependent claims 12, 13, 14, 16 and 17

Examination of new dependent claims 12, 13, 14, 16 and 17 would be appreciated.

C.5. New independent claims 15

New independent claim 15 has been drafted to include the limitations of independent claim 1 and dependent claims 6, 7 and 8.

New independent claim 15 further includes the limitation that the present process is a petroleum independent process. Claim 15 requires the glycerol to have been produced by one of three petroleum independent processes.

Examination of new independent claim 15 would be appreciated.

C.6. Prosecution compared to later filed and already granted U.S. Patent No. 7,396,962

The present case claims the benefit of JP 2004-024181 filed January 30, 2004. The International Filing Date of

10/585,793

the present case is January 28, 2005. Of course, the present case has yet to be allowed.

Dubois et al. filed their U.S. Provisional Patent Application 60/689,395 on June 10, 2005. The Dubois et al. provisional application later matured into the Dubois et al. U.S. Patent No. 7,396,962 issued July 8, 2008.

The Dubois et al. patent has one independent claim, reproduced below:

1. Process for manufacturing acrolein by dehydration of glycerol in the presence of an amount of molecular oxygen chosen so as to be outside the flammability range at any point in the process.

The title of the Dubois et al. patent is Process For Dehydrating Glycerol To Acrolein.

The Dubois et al. Notice of Allowability (three pages, Attachment A) provides that:

The following is an examiner's statement of reasons for allowance: the claims are drawn to a process for preparing acrolein by dehydration of glycerol. The prior art of record teach such a process; however, the prior art fails to teach or suggest conducting such a process in the presence of molecular oxygen. Accordingly, the instant claims are allowed.

The Dubois et al. Notice of Allowability does not mention the flammability range. The Dubois et al. specification discusses the flammability range as follows in column 3, lines 39-53:

One subject of the present invention is thus a process for manufacturing acrolein by dehydration of glycerol in the presence of molecular oxygen. The molecular oxygen may be present in the form of air or in the form of a mixture of gases containing molecular oxygen. The amount of oxygen is chosen so as to be outside the flammability range at any point in the plant. From FIG. 4 of US patent application 2004/15012, the maximum oxygen content, in an acrolein/O₂/N₂ mixture is about 7%

10/585,793

by volume in order to be entirely outside the flammability range. The oxygen content in the process according to the invention will generally be chosen so as not to exceed 7% relative to the mixture of gases entering the reaction (mixture of glycerol/H₂O/oxygen/inert gases). Preferably, the oxygen content is less than 7% relative to the dry gas mixture leaving the reactor (mixture of acrolein/oxygen/inert gases).

The present specification discloses:

[0016] In addition, in the aforementioned gas phase oxidation reaction, it is preferable that: when a flow of the gas, formed by the aforementioned dehydration reaction, is passed through the reactor, oxygen gas is beforehand added to that gas to increase the oxygen concentration. By this operation, the reactivity of the oxidation reaction is enhanced, so that acrylic acid can be obtained in a higher yield. Specifically, it is preferable that the oxygen concentration in the gas being supplied to the reactor of the aforementioned gas phase oxidation reaction is controlled to not less than 2% by volume. Incidentally, if the oxygen concentration in the gas being supplied to the reactor of the aforementioned gas phase oxidation reaction is too much high, then there occurs an unfavorable possibility that it may fall within the combustion range to thus involve risks of such as explosion. Therefore it is preferable that the upper limit value of the oxygen concentration is appropriately determined so as to avoid the combustion range in consideration of such as the concentration of unreacted raw glycerol, contained in the gas formed by the dehydration reaction, and the reaction temperature. [Page 5, line 18 to page 6, line 4.]

[0030] An aqueous glycerol solution having a water content of 15% by weight was vaporized, and an oxygen-containing gas was added thereto. A flow of the resultant mixed gas (gas composition: glycerol 10% by volume, water 9% by volume, oxygen 6% by volume and nitrogen 75% by volume) was passed through the aforementioned first-step reaction tube and the aforementioned second-step reaction tube in sequence at a flow rate of 120 mL/min, and then the discharged gas was collected with a

(18450.DOC) (Amendment and Remarks--page 15 of 19)

10/585,793

collecting bottle containing water to obtain acrylic acid. Its yield was 55%. [Example 1, page 11, lines 5-11.]

[0034] An aqueous glycerol solution having a water content of 8% by weight was vaporized, and an oxygen-containing gas was added thereto. A flow of the resultant mixed gas (gas composition: glycerol 10% by volume, water 4% by volume, oxygen 6% by volume and nitrogen 80% by volume) was passed through the aforementioned reaction tube at a flow rate of 420 mL/min, and then the discharged gas was collected with a collecting bottle containing water to obtain acrylic acid. Its yield was 65%. [Example 3, page 12, lines 13-18.]

[0036] An aqueous glycerol solution having a water content of 2% by weight was vaporized, and an oxygen-containing gas was added thereto. A flow of the resultant mixed gas (gas composition: glycerol 10% by volume, water 1% by volume, oxygen 6% by volume and nitrogen 83% by volume) was passed through the aforementioned reaction tube at a flow rate of 420 mL/min, and then the discharged gas was collected with a collecting bottle containing water to obtain acrylic acid. Its yield was 57%. [Example 4, page 12, line 25 to page 13, line 3.]

[0038] An aqueous glycerol solution having a water content of 9% by weight was vaporized, and an oxygen-containing gas was added thereto. A flow of the resultant mixed gas (gas composition: glycerol 10% by volume, water 5% by volume, oxygen 6% by volume and nitrogen 79% by volume) was passed through the aforementioned reaction tube at a flow rate of 420 mL/min, and then the discharged gas was collected with a collecting bottle containing water to obtain acrylic acid. Its yield was 58%. [Example 5, page 13, lines 9-14.]

The first and second steps of the present case generally are:

1. glycerol to a gaseous reaction product (acrolein).
2. gaseous reaction product (acrolein) to acrylic acid.

Claims 1 and 7 of the present application presently read:

10/585,793

1. (previously presented) A process for producing acrylic acid, comprising the steps of:

- vaporizing a raw material comprising an aqueous glycerol solution to generate a first gas, wherein said aqueous glycerol solution has a water content of not more than 20% by weight;
- applying a dehydration reaction to glycerol in a gas phase that includes said first gas; and then
- applying a gas phase oxidation reaction to a gaseous reaction product [acrolein] formed by the dehydration reaction to obtain said acrylic acid.

7. (previously presented) The process for producing acrylic acid according to claim 1, wherein the gas phase in which the dehydration reaction is conducted is a mixed gas comprising glycerol, water vapor and oxygen.

Claim 7 of the present application recites that the dehydration reaction of glycerol is conducted in a mixed gas comprising glycerol, water vapor and oxygen. Therefrom, it is clear that the dehydration reaction of glycerol as defined in this claim 7 is, like the Dubois et al. patent, conducted "in the presence of molecular oxygen."

Therefore, for the same reasoning as applied to the Dubois et al. patent in the Dubois et al. Notice of Allowability, it is respectfully submitted that dependent claim 7 of the present case is allowable.

D. Housekeeping matters

D.1. Period For Reply

A shortened statutory period for reply was set to expire three months from the mailing date of the Office Action of September 12, 2008. September 12, 2008 plus three months is December 12, 2008. This Amendment and Remarks is being filed on or before Friday, December 12, 2008 and may

10/585,793

be being filed on or before Wednesday, November 12, 2008.

D.2. Status

The Office Action of September 12, 2008 was made final.

D.3. Disposition Of Claims

Claims 1 and 3-9, and 11-17 are pending.

D.4. Application Papers

This case includes no drawings.

D.5. Priority under 35 U.S.C. §§ 119 and 120

Acknowledgement of the claim for foreign priority was made in The Office Action dated May 24, 2007. This is appreciated.

D.6. Attachments

As the Examiner has indicated, applicant has filed one PTO-1449 form in this case. This form has been initialed, signed and returned. This is very much appreciated.

Consideration of the Information Disclosure Statement of September 9, 2008 would be appreciated.

Consideration of the Information Disclosure Statement submitted with the present Amendment and Remarks would be appreciated.

E. Summary

Applicant respectfully submits that the present application is now in condition for allowance. The Examiner is respectfully invited to make contact with the undersigned

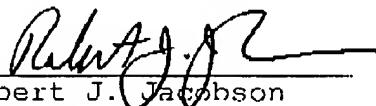
10/585,793

by telephone if such would advance prosecution of this case.

Respectfully submitted,

Date: 11-9-2008

Tel. No.: (651) 699-7900
Fax. No.: (651) 699-7901



Robert J. Jacobson
Reg. No. 32,419
650 Brimhall Street South
St. Paul, MN 55116-1511

(18450.DOC) (Amendment and Remarks--page 19 of 19)